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DIVISION OF ISOTOPE
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March 1970

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UC-23, Radioisotope and
Radiation Applications

PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPE DEVELOPMENT PROGRAMS
QUARTERLY REPORT: NOVEMBER - JANUARY 1970

Edited by W. F. Sheely

March 1970

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ABSTRACT

PROMETHIUM COMPATIBILITY

The experimental evaluation of $^{147}\text{Pm}_2\text{O}_3$ compatibility with capsule materials has been completed. The preparation of an unclassified document detailing the results of the complete $^{147}\text{Pm}_2\text{O}_3$ compatibility program is now in progress. The compatibility testing of various containment materials with samarium metal (stand-in for $^{147}\text{Pm}_2\text{O}_3$) was continued on schedule.

ENCAPSULATED WASTE MANAGEMENT CESIUM AS A PROCESS RADIATION SOURCE

Program discussions with DID resulted in redirection of the program toward improved design of waste containers for efficient use as irradiation sources. Reconsideration of the shielding analysis has shown that the quantitative evaluation of the effective source strength must be on the basis of target geometry as well as source geometry.

RADIATION CHARACTERISTICS OF FISSION PRODUCT RHODIUM AND PALLADIUM

Collection of data needed to obtain the half-life of $^{102\text{m}}\text{Rh}$ is continuing.

NONDESTRUCTIVE TESTING TECHNIQUES

NDT for Irradiation Sources

The current objective of this task is to develop a multi-element, ultrasonic transducer which will permit scanning of wide paths in large, flat surfaces (such as plate-type irradiation sources) for internal defects. The electronic circuitry

for signal generation, signal amplification, and transducer scanning has been designed, built, and tested. It functions well. Attention is now being directed toward the development of improved readout and display instrumentation.

Neutron Radiography with ^{252}Cf

Research in neutron radiography with small, non-reactor sources has been advanced by the development of a portable neutron device using ^{252}Cf . The use of ^{252}Cf in such a device has been confirmed by both analytical and experimental methods. A first generation neutron camera (NC-1) has been designed and built and is presently being evaluated. Preliminary results include neutron radiographs and flux measurements.

ADVANCED SOURCE DEVELOPMENT

Preparation of $^{241}\text{AmO}_2$ and UO_2 radiation sources was completed in this period. A wide variety of source designs were fabricated. Preliminary tests have demonstrated the feasibility of the sputtering process for deposition of radioisotope layers and isotope containment windows. Further process refinement is indicated.

EVALUATION OF ISOTOPE PRODUCTION IN POWER REACTORS

The ^{238}Pu production cost studies have continued along the lines of developing comparisons of neptunium irradiations in power reactors with irradiations in high flux irradiations. Because of a low-lying resonance peak in the ^{237}Np absorption cross section, variations in neutron temperature have a significant effect on target conversion rates and yields. Results using the BURN and TAIRCOST programs show that the optimum ^{238}Pu production costs are reduced 3 to 4% per 100 °C of increased neutron temperature.

RADIATION CHARACTERISTICS OF CIRCULATORY SUPPORT ISOTOPIC HEAT SOURCES

Work continues on improving the shielding and in-phantom dose predictions for the LASL-1 sources. The in-phantom dosimetry measurements of LASL-1 and ^{252}Cf in the Cornell dog-ball continues. In addition, calorimetric measurements of LASL-1 are nearly completed.

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INTRODUCTION

This is the thirteenth report of a series initiated in 1966 to provide detailed and timely information on the results of the isotopic fuels program and related research being conducted at the Pacific Northwest Laboratory by Battelle-Northwest for the U. S. Atomic Energy Commission. The previous reports of the series are BNWL-385, 466, 570, 680, 681, 781, 877, 924, 1010, 1085, 1177, and 1256.

This report covers the 3-month period, November 1969 through January 1970.

PROMETHIUM COMPATIBILITY

H. T. Fullam and J. H. Jarrett

The experimental evaluation of $^{147}\text{Pm}_2\text{O}_3$ compatibility with capsule materials has been completed. Electron beam microprobe analysis of two, 12-month, 1100 °C $^{147}\text{Pm}_2\text{O}_3$ test specimens was completed in December. No significant amount of $^{147}\text{Pm}_2\text{O}_3$ -capsule material reaction or inter-diffusion was indicated. The preparation of an unclassified document detailing the results of the complete $^{147}\text{Pm}_2\text{O}_3$ compatibility program is now in progress.

The compatibility testing of various containment materials with samarium metal (stand-in for ^{147}Pm metal) was continued. The heating of the 8800 hr, 400 °C and 700 °C test specimens was completed. Heating of the 4400 hr, 1000 °C test specimens also was completed. Metallography is now being performed on each specimen.

ENCAPSULATED WASTE MANAGEMENT CESIUM
AS A PROCESS RADIATION SOURCE

H. H. Van Tuyl

Program discussions were held with DID to determine the appropriate direction of the program with respect to cesium sources. Choice of encapsulation materials and dimensions of desired radiation sources were discussed. Waste Management selected 316L SS as one candidate material because of its prior use for encapsulating cesium chloride and its ease of fabrication. Hastelloy C was selected as the other candidate because of its expected corrosion resistance. Either material seems acceptable for a radiation source provided compatibility is demonstrated. Continuing compatibility studies conducted in the Waste Management Program show good results with both materials.

Optimization of the cesium container as a radiation source was discussed. Current Waste Management plans are for a container 2.25 in. OD by 23.8 in. long (a slight increase in length over the last few months). Total wall thickness is 0.204 in. Much of the cesium radiation is lost within the source, but the radiation which escapes from the source is still penetrating enough to be useful for most radiation source applications. An improved radiation source configuration of 1 in. diameter by 18 in. long was chosen previously for preliminary studies. Reconsideration of this approach resulted in a decision to couple cost studies with source optimization. Some of the items to be considered are efficiency of cesium utilization, need for additional facilities or capital equipment at Waste Management, effect on operating costs, shipping problems, use in existing or new irradiation facilities, and licensing problems.

Preliminary evaluation of proposed Waste Management facilities indicates that the number of capsules to be fabricated can be increased without requiring additional facilities or appreciable amounts of additional capital equipment. Estimates of the possible increase range from less than twofold to as much as fivefold, depending on assumed operating efficiency and ability to devise adequate holders and seals to permit filling many capsules simultaneously. The amount of increase cannot be determined conclusively without experimental work, but a twofold increase does not appear to be unreasonable. An increase in total operating costs is expected with any increase in the number of cesium containers, but unit costs are not expected to change greatly.

Efficiency of radiation use has been calculated as a function of source diameter and container thickness. Self absorption by the CsCl in the source was calculated on the basis of energy absorption (Table 1), using methods similar to those developed by Galanter of BNL. A source diameter of 2.5 in. results in transmission of 66% of the total gamma energy, which is 20% lower than the amount transmitted for a 1 in. diameter source. This shows the strong incentive for reducing source diameter. The storage container as currently envisioned is somewhat smaller (1.81 in. ID), but there is still an incentive for reducing diameter.

TABLE 1. Energy Absorption Within a CsCl Source

<u>Source Diameter, in.</u>	<u>Absorption, %</u>
1.0	17
1.5	23
2.0	29
2.5	34

Most of the attenuation of the primary radiation is due to Compton scattering. A secondary photon is emitted with an energy dependent on the angle of scatter and initial photon energy. For ^{137}Cs gammas, the average scattered energy is 0.409 MeV, so that only 0.253 MeV is actually lost in the first scattering. However, the scattered photons are less penetrating than the primary photons, so minimal scattering in both the source and container is desired, especially if thick targets are being irradiated. Optimization of source size will depend on target configuration in addition to encapsulation cost, pricing, and source efficiency.

Self absorption was evaluated with respect to energy absorption since this represents a compromise between attenuation of dose rate and attenuation of primary radiation. The effect of container thickness was evaluated for the extreme cases (Table 2) to show the extent of variation which must be considered.

TABLE 2. Attenuation of ^{137}Cs Radiation by Steel Container

<u>Container Thickness, mils</u>	<u>Attenuation of Primary, %</u>	<u>Attenuation of Dose Rate, %</u>
25	3.6	0.7
50	7.0	1.5
75	10	2
100	14	4
125	17	5
150	20	6
175	23	7
200	25	8

Neither basis is entirely satisfactory since the first basis is applicable only to deep penetration into a target and the second is applicable only at the surface of a target.

A reduction in container thickness to 100 mils total would result in more efficient use of the radiation, but the amount of improvement would depend on target geometry.

RADIATION CHARACTERISTICS OF FISSION PRODUCT
RHODIUM AND PALLADIUM

F. P. Roberts

The objectives of this study for the current fiscal year include the measurement of the half-life of ^{102m}Rh , measurement of the cross-section for the formation of ^{102m}Rh , and analysis of the ^{102m}Rh content of rhodium from spent power reactor fuel.

Efforts during this quarter have been directed at the half-life measurements. Samples of 13 year-old rhodium from the Hanford reactors have been highly purified and are being counted routinely to follow the decay of the ^{102m}Rh . Because of the extreme age of the rhodium in these samples no ^{102}Rh is present, and the change in the counting rate can be attributed to the decay of the ^{102m}Rh .

NONDESTRUCTIVE TESTINGNDT FOR IRRADIATION SOURCES

R. W. Steffens

NDT methods and device development continued with the evaluation of multi-element ultrasonic test concept instrumentation. The ultrasonic signal generation, signal amplification, and transducer scanning circuits were found to function well. The performance of these circuits permitted the establishment of the practicality of a CRT readout of the multi-element signals. The successful demonstration of the CRT readout display warranted the initiation of prototype signal synthesis and readout circuitry development.

The pulser, preamplifier, and electronic system clock were fabricated into a prototype instrument for evaluation with the multi-element ultrasonic transducers and the recently fabricated transducer alignment mechanisms. The integrated effect of the circuits and mechanisms produced good signal-to-noise ratios at the receive transducer which are compatible with signal synthesis requirements. The acquisition of good signal-to-noise ratios establishes the practicality of the multi-element transducer test concept and justifies initiation of signal synthesis and readout circuitry development.

The ability to obtain a CRT readout of the multi-element transducer signals was demonstrated by interfacing the present prototype multi-element signal generation and scanning instruments with the UT-40 signal synthesizing circuits and an oscilloscope. The results showed the feasibility of using a Z-axis modulated (beam intensification) CRT for displaying the multi-element data. An oscillogram of test results obtained using a "cold" ^{60}Co irradiation source with reference drill holes is shown in Figure 1. Note, the readout consists of a dual intensity matrix of dots which is 32 dots across. Each

FIGURE 1. Intensified CRT Readout of Multi-element Transducer Signals and Associated Equipment

dot in the 32-dot row corresponds to an individual element of the multi-element transducer and the spacing between the 32-dot rows represents translation of the transducer down the length of the irradiation strip. The three areas of brightly intensified dots represent the detection of the drill hole references and the less intense dots represent normal source conditions. The signal synthesis circuit presently under development will improve the dot contrast considerably and will provide more reliable readout data to the oscilloscope than is presently available in the UT-40 test console.

The prototype multi-element concept instrumentation is expected to be completed around March 31, 1970. Application of the multi-element test techniques will then be demonstrated, and the instrument-application technology will be published.

NEUTRON RADIOGRAPHY WITH ^{252}Cf

J. L. Cason

The program initiated at the PNL for the development of a portable device for neutron radiography is well into the third and fourth phases of work. Phases I, II, and first part of Phase III were completed during the last quarter. The work involved an analytical investigation, source acquisition, and initial experiments. (Ref. BNWL-177)

The initial part of Phase III included the construction of an experimental device which provided variable collimation, moderation, and source positioning. Neutron radiographs were made using this device which demonstrated the usefulness of a portable unit for neutron radiography.

Recent accomplishments under Phase IV include

- Definition of optimum moderator conditions.
- Evaluation of imaging technique (limited to 1 specific case).

- Evaluation of shielding materials.
- Use of metal hydride as a moderator.
- Definition of design requirements for portable device.
- First generation design of portable device.
- Fabrication of portable device.
- Initial testing, mechanics, etc., of device.
- Preliminary radiographic and flux measurements.

Efforts under way at present include the following:

- Complete testing of the physical properties of neutron camera NC-1 construction material.
- Preparation of report providing design and fabrication details of NC-1.
- Further evaluation of NC-1 radiographic capabilities.

Discussion

A factor that has hindered the growth of neutron radiography application is the absence of a suitable portable source that would provide a high intensity of neutrons over a reasonable period of time. The development of a portable, low cost, high thermal flux source will aid in many areas of applications where large immobile devices such as reactors and generators are not suitable.

Extensive research has been performed in search of a solution to this problem. The most apparent approach was the development of a radioactive isotope source. Research with Sb-Be, Po-Be, Am-Be and other such (αn) or (γn) reaction neutron sources has shown that radiography with isotopes is feasible. However, shielding requirements and short half-lives for these types of sources were undesirable.

Recent developments using ^{252}Cf have shown that it can be used in a portable device for thermal neutron radiography. The physical properties of ^{252}Cf which make it extremely attractive as a radiographic source are:

Effective half-life	2.65 years
Average neutron energy	2.34 MeV
Neutron yield/gram	2.34×10^{12} neutrons/sec.

A 1-gram source when properly moderated and collimated could produce a thermal flux of about 1×10^7 nv at the object position. This would compete with most reactors for applications in neutron radiography.

Experimental Results

A 268 μg ^{252}Cf source has been used in an experimental device (Figure 2a and 2b) to produce neutron radiographs. Using optimum moderator characteristics as given in Figure 3, the experimental camera can produce a thermal flux of collimated neutrons of 1×10^4 nv at the object position. A radiograph taken with this experimental arrangement is shown in Figure 4. The direct technique was used, as shown in Figure 5. The direct and indirect techniques are compared in Figure 5. Imaging foils useful for neutron radiography are listed in Table 3.

FIGURE 2a. Experimental Neutron Radiography Source Camera - Specifications: 10 in. by 10 in. by 12 in. long; Weight, ~50 lb

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FIGURE 2b. Experimental Neutron Radiography Source Camera -
Located in Hot-Cell

FIGURE 3. Calculated Thermal Flux for Various Moderators

- (a) BOOBY TRAPPED WITH PLASTIC EXPLOSIVE (g)
 (b) LOADED WITH CHEMICAL TO SIMULATE HEROIN (h)
 (c) CADMIUM TEST PIECE

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FIGURE 4. Neutron Radiograph - Ball Point Pens

DIRECT EXPOSURE METHOD

TRANSFER EXPOSURE METHOD

FIGURE 5. Neutron Radiography - Imaging Techniques

TABLE 3. Characteristics of Several Neutron Converter Materials

Material	Isotope Involved in Reaction	Relative Natural Abundance, %	Cross-Section for Reaction (thermal neutrons, velocity = 2,200 m/sec), barns	Reaction	Half-Life s = sec m = min h = hours d = days
Lithium	Lithium-6	7.52	910	${}^6\text{Li}(n,\alpha){}^3\text{H}$	
Boron	Boron-10	18.8	3,770	${}^{10}\text{B}(n,\alpha){}^7\text{Li}$	
Rhodium	Rhodium-103	100	12	${}^{103}\text{Rh}(n){}^{104\text{m}}\text{Rh}$	4.5 m
			140	${}^{103}\text{Rh}(n){}^{104}\text{Rh}$	44 s
Silver	Silver-107	51.35	44	${}^{107}\text{Ag}(n){}^{108}\text{Ag}$	2.3 m
	Silver-109	48.65	2.8	${}^{109}\text{Ag}(n){}^{110\text{m}}\text{Ag}$	270 d
			110	${}^{109}\text{Ag}(n){}^{110}\text{Ag}$	24.2 s
Cadmium	Cadmium-113	12.26	20,000	${}^{113}\text{Cd}(n,\gamma){}^{114}\text{Cd}$	
Indium	Indium-115	95.77	155	${}^{115}\text{In}(n){}^{116\text{m}}\text{In}$	54.1 m
			52	${}^{115}\text{In}(n){}^{116}\text{In}$	13 s
Samarium	Samarium-149	13.8	40,800	${}^{149}\text{Sm}(n,\gamma){}^{150}\text{Sm}$	
	Samarium-152	26.8	140	${}^{152}\text{Sm}(n){}^{153}\text{Sm}$	47 h
Gadolinium	Gadolinium-155	14.73	61,000	${}^{155}\text{Gd}(n,\gamma){}^{156}\text{Gd}$	
	Gadolinium-157	15.68	240,000	${}^{157}\text{Gd}(n,\gamma){}^{158}\text{Gd}$	
Dysprosium	Dysprosium-164	28.1	2,000	${}^{164}\text{Dy}(n){}^{165\text{m}}\text{Dy}$	1.25m
			500	${}^{164}\text{Dy}(n){}^{165}\text{Dy}$	140 m
Gold	Gold-197	100	96	${}^{197}\text{Au}(n){}^{198}\text{Au}$	2.7 d

Paraffin and ZrH_2 were selected for an experimental comparison as the most likely moderators to be used in a portable device.

Table 4 compares the moderating ratios of several standard moderators. The calculated flux values for the various optimum moderator cases are given in Table 5 for comparison to the calculated results. Hydrogen density of the ZrH_2 used in this experiment is 7.8×10^{22} hydrogen atoms/cm³ as compared to 6.7×10^{22} hydrogen atoms/cm³ for H_2O at STP. As would be expected the ZrH_2 is the more efficient moderator.

Experimental testing with the neutron camera included neutron shielding evaluations. It was found that a 60% by weight mixture of boron carbide powder (B_4C) in epoxy resin was the most effective against fast neutrons. The epoxy resin provided enough hydrogen to moderate the fast neutrons and the B_4C captured the thermal neutrons. This shield was more effective than CH_2 , H_2O or any of the metal hydrides tested, ZrH_2 and TiH_2 .

TABLE 4. Moderating Ratios for Various Materials

<u>Material</u>	<u>Slowing Down Power $\xi\Sigma_s$</u>	<u>$\Sigma_a(\text{CM}^{-1})$</u>	<u>Moderating Ratio</u>
Be	0.16	0.0011	150
C (Graphite)	0.063	0.00037	170
Light H ₂ O	1.50	0.022	69
Heavy H ₂ O (99.17% D ₂ O)	0.18	85×10^{-6}	2100
Polystyrene (CH) _n	0.80	0.013	62
ZrH _{1.94}	1.54	0.030	51

TABLE 5. Flux Values Using Paraffin and ZrH₂ Moderators

	<u>Calculated Thermal Flux Optimum Moderator Case</u>	<u>Measured Thermal Flux (In Foil Activation) Optimum Moderator Case</u>
Paraffin	6.7×10^5 nv	6.6×10^5 nv
ZrH ₂	1.2×10^6 nv	1.3×10^6 nv

Neutron Camera Design

The evaluations with the experimental device provided the requirements for the design and fabrication of a portable, neutron camera (NC-1). The camera (a), pictured in Figure 6, has the following features:

- Completely contained unit, providing a safe and an expose position for the ²⁵²Cf.
- Variable moderation plus heating and cooling capability for the metal hydrides.
- Variable divergent beam collimator.

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FIGURE 6. Neutron Radiography Camera NC-1

- Fire proof.
- Highly impact resistant.
- Remote operation (b)
- Remote monitoring of source position.
- Remote monitoring of neutron level near device (d).
- Manual device for storing the source should the automatic device malfunction.
- Relatively light weight ~100 pounds.
- Time Recorder for exposures - integrated neutron dose (e).

The internal shielding conditions of the device provide a surface dose rate of only 200 mR/hr neutrons and 50 mR/hr gamma for a 268 μg ^{252}Cf source. The neutron beam is highly directional with a measured flux at the moderator surface and object position 18 in. from the source of 1.3×10^6 nv thermal and 1.1×10^4 nv thermal, respectively.

These conditions permit high quality neutron radiographs to be taken in times ranging from a few minutes to several hr. The time required for radiography directly depends upon the imaging technique and properties of the object being radiographed.

A $^6\text{LiF Zn S(Ag)}$ scintillation screen (direct technique) can produce a neutron radiograph in about 1 min whereas Dy (indirect technique) foils require 3 hr activation to produce a transfer image. Figure 7a is a radiograph taken with the neutron camera in an exposure time of about 1 hr and can be compared to an X-ray of the same object. The Gd direct technique was used with standard X-ray film. The same radiograph could have been made using a $^6\text{LiF Zn S(Ag)}$ screen in about 1 min due to their relative speed differences of 70:1 for the screen as compared to the Gd foil.

A complete evaluation of the neutron camera's characteristics and capabilities is planned as the next step. This evaluation includes:

- Physical properties of fiberglass construction material, i.e., tensile strength, compressibility, thermal conductivity, etc.
- Conformity to Federal Shipping Regulations.
- Radiographic capabilities and limitations.

a. Neutron Radiograph

b. X-ray Radiograph

Key to Objects:

I. Top to bottom small squares.

0.010 in. Cu	0.010 in. Au
0.060 in. Paraffin	0.010 in. In
0.040 in. Paraffin	0.020 in. In
0.020 in. Paraffin	0.030 in. In
0.020 in. Cd	0.015 in. Pb
0.040 in. Cd	0.030 in. Pb
0.060 in. Cd	0.045 in. Pb

II. Test strip with drilled holes.

III. Booby trapped M-16 round.
Tracer loaded M-16.
Standard loaded M-16.

FIGURE 7. Neutron and X-ray Radiograph Comparison

Publications and Presentations

J. L. Cason and C. B. Shaw. "Neutron Radiography with Cf^{252} ," ANS Transactions 12, No. 2, p. 464-4. November 1969.

J. L. Cason. "Californium - 252, for Neutron Radiography," Californium - 252 Progress - Its Use and Market Potential, No. 1, p. 7, October 1969. SRL. Savannah River Operations, USAEC.

J. L. Cason. "Neutron Radiography with Cf^{252} in Forensic Sciences," invited paper, 7th Annual Symposium of Forensic Science, Armed Forces Institute of Pathology, Washington, D.C. October 1969.

J. L. Cason. "Californium - 252, for Neutron Radiography," Californium - 252 Progress - Its Use and Market Potential, No. 2, February 1970. SRL. Savannah River Operations, USAEC.

Papers Accepted for Presentation

C. B. Shaw and J. L. Cason. "Portable Neutron Radiographic Camera Cf^{252} ." Spring Meeting ASNT, March 1970.

Reports in Progress

Design and Fabrication of a Portable Device for Neutron Radiography with $\text{Cf}-252$.

ADVANCED ALPHA SOURCE DEVELOPMENT

J. E. Hansen

Work in this area is divided into four technical areas:

- Determination of substrate criteria for optimum deposit-substrate bonding.
- Determination of window deposit fabrication parameters for optimum deposit bonding and integrity.
- Determination of preferred techniques for depositing the isotope source material on the substrate.
- Theoretical analyses to determine preferred material types and structures for specific applications to provide conceptual source designs offering maximum isotope containment, and to provide a continuing basis for experimentation.

Evaluation of the deposit-substrate bonding relationship is essentially complete within the present scope of this program. Results of experimentation in this area were reported previously (reference BNWL-1256).

Work in this period included start-up of the rf-sputtering system in a glovebox, and preparation of $^{241}\text{AmO}_2$ and UO_2 containing radiation sources. The sputtering system installation is shown on Figure 8. The sputtering system has been operated for 150 to 200 hr in the glovebox.

The following radiation sources have been prepared:

- UO_2 - Cu cermet substrate
 - Cu window only
 - Cu window with Pt overlay
- UO_2 - Cu cermet sputtered onto solid SS substrate
 - Cu window only
- UO_2 - Cu cermet sputtered onto solid Cu substrate
 - Cu window only

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FIGURE 8. View of the rf-Sputtering System Within a Glovebox

- AmO_2 - Cu cermet substrate
 - Cu window only
 - SS window only
- AmO_2 - Cu cermet sputtered onto solid SS substrate
 - SS window only
- AmO_2 - Cu cermet sputtered onto solid Cu substrate
 - Cu window with SS overlay

Preparation of these sources has demonstrated the ability of the rf-sputtering process to deposit radioisotopes and containment windows.

Figure 9 illustrates a sputtering target typical of the UO_2 -Cu and AmO_2 -Cu type. These targets are prepared by compaction of the cermets, followed by cementing the resulting tiles on a Cu backing.

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FIGURE 9. Typical AmO_2 -Cu or UO_2 -Cu Cermet Sputtering Target

The sources prepared have been subjected to various tests of a preliminary nature. Test results are presented on Table 6. These tests have identified areas where source design and fabrication parameters require refinement. The last sources prepared (AmO_2 -Cu sputtered onto Cu with Cu and SS windows) incorporate several refinements which are expected to result in high quality alpha-photon sources. Testing of these sources will be performed in the next period.

TABLE 6. Preliminary Tests on Sputtered Radiation Sources

Source Description	Test	Test Results and Comment
UO ₂ -Cu cermet substrate with $\sim 1 \mu$ copper window	Smear	Free of contamination after cleaning with soap and water
	Ultrasonic vibration for 30 min in water	Window failure in areas where UO ₂ was agglomerated. Problem of heterogeneous cermet substrate and contaminated (oxidized) copper powders.
UO ₂ -Cu cermet substrate with $\sim 1 \mu$ copper window and $< 0.1 \mu$ Pt overlay	Smear	Free of contamination.
	Exposure to dilute HCl	Corroded window. Platinum overlay apparently too thin to protect copper from acid.
UO ₂ -Cu cermet sputtered onto solid copper substrate with $\sim 2 \mu$ copper window	Smear	Free of contamination.
	Ultrasonic vibration for 25 min in water at room temp	No discernible failure.
	Ultrasonic vibration for 18 hr in water at 120-140 °F	Slight pitting of copper window observed with some loss of activity to water.

TABLE 6. (contd)

Source Description	Test	Test Results and Comment
UO ₂ -Cu cermet sputtered onto solid stainless steel substrate with ~2 μ copper window	Smear Other	Free of contamination. These large (>3-in. diameter) sources were used for visual examination of deposit uniformity and for testing of resistance to scratching. A rather abusive effort was required to scratch through the sputtered layers.
AmO ₂ -Cu cermet substrate with <1 μ copper window	Smear Ultrasonic vibration for 20 min in water	Free of contamination. Window failure due to inadequate thickness of deposit over relatively rough surface of cermet type substrate.
AmO ₂ -Cu cermet substrate with <1 μ stainless steel window	Smear Ultrasonic vibration for 20 min in water	Free of contamination. Localized window failure. Problems of heterogeneous cermet substrate, oxidized copper powders, and too thin window.
AmO ₂ -Cu cermet sputtered onto solid stainless steel substrate, covered with <3 μ stainless steel window	Smear (quite abusive) Boiling for 15 min in water Ultrasonic vibration for 30 min in water Soak in water for ~70 hr	Free of contamination. No discernible failure. (Examined by smear and microscope.) No discernible failure. Both window and AmO ₂ -Cu deposit unbonded from substrate as integral film. Problem thought to be one of substrate preparation and/or presence of copper in isotope layer.

TABLE 6. (contd)

<u>Source Description</u>	<u>Test</u>	<u>Test Results and Comment</u>
AmO ₂ -Cu cermet sputtered onto solid copper substrate, covered with ~5 μ copper followed by <3 μ stainless steel	Smear	Free of contamination.
	Heating in flowing air at 400 °F for 15 min	No discernible failure.
	Heating in flowing air from 250-925 °C in 2 hr	Visual effects noted on source surface but containment retained.
	Other	Testing is yet to be performed.

EVALUATION OF ISOTOPE PRODUCTION IN POWER REACTORS

R. W. McKee and S. E. Nunn

PLUTONIUM-238 COST STUDIES - THE EFFECT OF NEUTRON FLUX AND COST

The ^{238}Pu production cost studies have continued along the lines of developing comparisons of neptunium irradiations in power reactors with irradiations in high flux reactors.

Results reported in the previous quarterly report⁽¹⁾ showed that for a fixed set of cost inputs, there are only minor differences in optimized ^{238}Pu production costs over a range of neutron fluxes from 2.5×10^{13} n/cm²/sec to 1×10^{14} n/cm²/sec. However, these results were based on a constant neutron temperature of 400 °C which is a typical power reactor neutron temperature but is not necessarily representative of a high flux reactor. For example, the bulk moderator temperature for the heavy water moderated Savannah River high flux demonstration⁽²⁾ was reported to be 67 °C, indicating average thermal neutron temperatures on the order of 100 °C.

Because of a low-lying resonance peak in the ^{237}Np absorption cross section, variations in neutron temperatures have a significant effect on target conversion rates and yields. To identify the magnitude of the effects of neutron temperature variations on ^{238}Pu production costs, a series of TAIRCOST program calculations were carried out. The target conversion rate and yield data used in the calculations were obtained with the BURN program.

Results of these calculations show that the optimum ^{238}Pu production costs are reduced 3 to 4% per 100 °C of increased neutron temperature. This relationship was found to hold true over a neutron temperature range from 200 to 600 °C, over neutron flux levels from 2.5×10^{13} to 1×10^{14} n/cm²/sec and over a broad range of cost assumptions including neptunium

values from \$50 to \$100 per gram, neutron costs from \$2700 to \$10,000 per gram and the high and low processing cost range used in previous comparisons.⁽¹⁾ Thus, high neutron temperature provides a significant cost advantage for ^{238}Pu production.

REFERENCES

1. Pacific Northwest Laboratory Division of Isotope Development Programs Quarterly Report: August-October 1969, BNWL-1256. Battelle-Northwest, Richland, Washington, August 1969.
2. J. L. Crandall. The Savannah River High Flux Demonstration, DP-999. E. I. du Pont de Nemours and Company, Aiken, South Carolina, June 1965.

RADIATION CHARACTERISTICS OF CIRCULATORY
SUPPORT ISOTOPIC HEAT SOURCES

F. T. Cross and J. C. Sheppard

SHIELDING AND IN-PHANTOM DOSE PREDICTION

The Battelle-Monte Carlo code, BMC, and Battelle's nuclear data, ENDF/B, were used to calculate fission neutron fluence-to-dose conversion factors for a ^{238}Pu source implanted centrally in a cylindrical tissue phantom, 35 cm diameter by 50 cm height. The final conclusions are:

- (a) Within the errors of the calculations the fluence-to-dose conversion factors are identical for the axial and radial directions.
- (b) The proton recoil fluence-to-dose conversion factors appear insensitive to the nature of the source and phantom geometry. The results of others for an external beam incident on a slab or cylinder of tissue are in essential agreement with those for a point source placed within a cylinder of tissue. Because the proton recoils are the largest contributor to the neutron dose in the cylindrical phantom, they largely determine the fluence-to-dose conversion factors.
- (c) The heavy ion recoil dose, which contributes less than 10% of the total dose, is about 30% greater than that calculated by Irving, Alsmiller and Moran of ORNL. The discrepancy can be attributed to differences in the scattering cross sections used in the two calculations.
- (d) As noted by others, the $\text{H}(n,\gamma)\text{D}$ fluence-to-dose conversion factor is quite sensitive to the phantom geometry. However, the contribution of these photons to the total neutron dose is not appreciable in the cylindrical phantom.

- (e) The total fluence-to-dose conversion factor curve is slightly convex, but is in essential agreement with the data of Irving, Alsmiller and Moran. Furthermore, doses calculated with QAD, which uses the moments method for an infinite medium, agree quite well with the ORNL calculations of Irving, Alsmiller, and Moran.
- (f) Neutron doses predicted by QAD are within 20% of the ORNL calculations out to about 20 cm of tissue. The agreement could be better if the neutron removal cross-sections and fluence-to-dose conversion factors are adjusted.

In addition, neutron doses are being calculated using the latest neutron fluence-to-dose conversion factors from ORNL (those compiled by Ritts, Solomito, and Stevens).

Tissue dose rates were also calculated for LASL-1 gammas in the source axial and perpendicular bisecting planes. The following input data were used in the QAD codes:

- Dimensions - taken from the LASL source and Battelle heat exchanger drawings.
- Gamma Abundances - taken from the most recent UCRL data (Lederer, Asaro and Perlman).
- Source Intensity - calculated for an assumed 31.0 watt source.
- Attenuation Coefficients - taken from LASL-3753 (Storm and Israel).
- Dose Buildup Factors - taken from the QAD Library.
- Flux to Dose Conversion Factors - taken from NBS Handbook 85.

Although the comparison of these calculations with the measurements is not complete, the following observations apply:

- If a weighted buildup factor is used (H_2O , Fe, and Pb were used in the calculations), the calculated dose-rates, although all were high, were within 20% of the measurements from 2 to 20 cm in tissue in the bisecting plane.
- The agreement along the axis was less good.
- The proper manner for weighting the dose calculations has not been thoroughly examined.
- The absolute heat output of LASL-1 has not yet been verified.
- Although the latest data of UCRL is assumed correct, there is still disagreement in the literature on gamma abundances.

IN-PHANTOM DOSIMETRY MEASUREMENTS

Calorimetric measurements, to determine the heat output of LASL-1, continue. The analysis of the Cornell ^{252}Cf dog-ball data is nearly completed and a report will be issued shortly. Generally, the measurements confirm the expected values discussed during the Cornell meeting in October and those reported to Cornell in correspondence.

The in-phantom measurements and comparisons with the predictions with LASL-1 continue.

^{146}Pm BURNOUT BY NEUTRON RADIATION

J. C. Sheppard

The ^{148m}Pm content of the Savannah River irradiated ^{147}Pm targets has now decayed to a level where analysis for ^{146}Pm , ^{147}Pm , and ^{148m}Pm can be performed and the $^{146}\text{Pm}/^{147}\text{Pm}$ ratios determined. The two Savannah River targets have been dissolved in preparation for the analysis, and the counting systems are being calibrated so that the samples can be counted in late January. $^{146}\text{Pm}/^{147}\text{Pm}$ ratios will be determined shortly after that time.

BNWL-1308-1

A document describing the Hanford ^{146}Pm burnout experiments, BNWL-1270, has been written and issued.

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